

Optical alignment and polarization conversion of neutral exciton spin in individual InAs/GaAs quantum dots

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(Dated: February 3, 2008)

We investigate exciton spin memory in individual InAs/GaAs self-assembled quantum dots via optical alignment and conversion of exciton polarization in a magnetic field. Quasiresonant phonon-assisted excitation is successfully employed to define the initial spin polarization of neutral excitons. The conservation of the linear polarization generated along the bright exciton eigenaxes of up to 90% and the conversion from circular- to linear polarization of up to 47% both demonstrate a very long spin relaxation time with respect to the radiative lifetime. Results are quantitatively compared with a model of pseudo-spin 1/2 including heavy-to-light hole mixing.

PACS numbers: 72.25.Fe, 72.25.Rb, 78.67.Hc, 73.63.Kv

Since the first proposal of using nanostructures for quantum computation [1] the semiconductor quantum dots (QDs) have been attracting a lot of attention. In such a system, quantum information can be stored and manipulated by using the degrees of freedom of a single or a few charge carriers, provided the coupling to the environment does not destroy the coherence. Single spin manipulation in a QD has been recently achieved by electron spin resonance [2, 3] and time-resolved Faraday or Kerr rotation [4, 5, 6], evidencing unequivocally the long coherence time of electron spin. In contrast, the spin coherence of a dipole-active neutral exciton (an electron hole pair) has been recently questioned in Ref. [7]: from rather indirect observations the authors conclude about a fast exciton spin relaxation (<100 ps) in self-assembled InAs/GaAs quantum dots. In this system, the bright excitons form a two-level system which can be considered as a pseudo-spin 1/2. In an ideal (cylindrical) QD, the eigenstates $|\pm 1\rangle$ are coupled to σ^\pm circularly polarized photons, but in actual QDs some symmetry reduction generally lifts the exciton degeneracy between two linearly-polarized states $|x\rangle$ and $|y\rangle$ by the fine structure splitting $\delta_1 \equiv \hbar\Omega_{exc.}$ (see Fig. 1(b)). Yet, the polarization quenching of these states was already demonstrated for excitons in an ensemble of InAs QDs under resonant excitation [8]. Investigating the longitudinal relaxation of single exciton spin by a straightforward method is thus highly demanded to elucidate the issue raised by Ref. [7].

In this work, we report on close to 100% optical alignment of the exciton dipole in individual quantum dots and close to 50% (theoretical maximum) conversion of polarization from *circular* to *linear* in a longitudinal magnetic field. These observations demonstrate that the exciton spin relaxation is quite negligible on the timescale of the exciton lifetime (~ 1 ns) for all the quantization directions defined by the effective magnetic field experienced by the quantum dot.

The sample was grown by molecular beam epitaxy on a

semi-insulating GaAs [001] substrate. It contains a vertical field effect structure with a single layer of InAs/GaAs quantum dots (see Fig. 1(a)). The detailed description of the structure can be found in Ref. [9]. Photoluminescence (PL) of neutral excitons was observed in the bias range $[-0.1 \text{ V}, 0.25 \text{ V}]$ so that for all the experiments reported here, the voltage was fixed at $+0.2 \text{ V}$. The PL excitation and collection were performed through the semitransparent part of the top Schottky gate.

The μ -PL spectroscopy of individual InAs QDs was performed using a split-coil magneto-optical cryostat. A 2 mm focal length aspheric lens (N.A. 0.5) was used to focus the excitation beam from a cw Ti:Sapphire laser and to collect the PL from the sample, while the relative positioning in all three directions was ensured by AttocubeTM piezo-motors with a precision of $\sim 0.5 \mu\text{m}$. This very compact microscope was immersed in the pumped liquid helium bath of the cryostat insert. All measurements were performed at low temperature ($T=2 \text{ K}$) and the magnetic field was applied parallel to the optical axis (Faraday configuration). The PL was dispersed using a 0.6 m-focal length *double* spectrometer and detected using a Nitrogen-cooled CCD array camera. A Babinet-Soleil compensator was used to correct for the residual birefringence of the cube beam splitter which separates the excitation and collection beams. The resulting extinction ratio was below 0.01 for both circular and linear cross-polarizations [10].

A prerequisite for the study of the exciton spin relaxation in QDs, is the ability to create a coherent superposition or a certain mixed state of the bright exciton states $|\pm 1\rangle$. To achieve this we used the quasiresonant 1 LO-phonon-assisted excitation (see Fig. 1(b)) which is compatible with standard cw PL spectroscopy of individual QDs. In InAs/GaAs quantum dots a marked resonance in the excitation spectrum occurs at $\sim 35 \text{ meV}$ above the ground state transition, i.e. between the GaAs LO- and TO-phonon features [9, 11, 12, 13]. Since the elec-

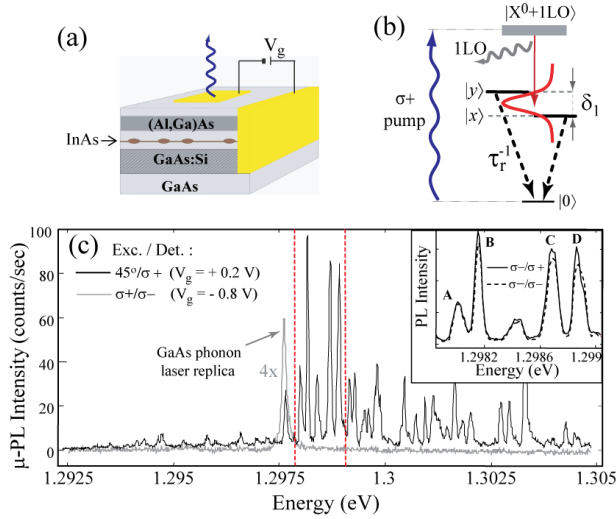


FIG. 1: (a) Sample structure of InAs/GaAs QDs used for controlling exciton charge state by an electric field. (b) Scheme of 1 LO-phonon-assisted excitation. (c) μ -PL spectra at quasi-resonant excitation for two different voltages. Dark line – neutral excitons observed at +0.2 V (45° excitation, $\sigma+$ detection), gray line – bulk GaAs Raman scattering obtained at -0.8 V in $\sigma+/\sigma-$ configuration. Inset: zoom of the region defined by the dashed lines bringing out 4 excitonic lines (A, B, C, D) with vanishing circular polarization under σ^+ excitation.

tron phonon interaction is essentially spin independent, this technique enables in principle to initialize the exciton spin state as performed in quantum wells [14], or more recently in self-assembled CdSe/ZnSe QDs [15, 16, 17] and InAs/GaAs QDs [18]. Creating a coherent state under these conditions requires that the exciton splitting δ_1 be not much greater than the exciton-phonon (or polaron) linewidth. Since the latter is determined by the fast decay (below 10 ps) of the LO-phonon part into acoustical phonons [19] this generally occurs, and indeed coherent exciton states have been evidenced by quantum beat measurements both in CdSe quantum dots [15] and InAs quantum dots [18]. We therefore consider in the following such a coherent state is generated, although our cw experiments did not enable us to probe the exciton coherence.

As illustrated in Fig. 1(c), the weak LO-phonon Raman scattering of bulk GaAs could be identified at exactly $\hbar\omega_{LO}=36.6$ meV below the laser energy in the cross-circular configuration of the excitation and detection polarizations and in an electric field of $F_z \sim 100$ kV/cm such that the PL signal from QDs is completely suppressed. At a positive bias (0.2 V) a few lines corresponding to neutral excitons are observed above the GaAs phonon replica of the laser which is likely due to the coupling with LO-phonons from InGaAs alloy of slightly lower energy [11]. In the rest of the paper we focus on 4 specific

Exciton	A	B	C	D
FSS δ_1 (μ eV)	26	16	29	50
Eigenaxis angle φ_0 ($^\circ$)	69	83	121	155

TABLE I: Parameters of fine structure splitting of all four selected excitons.

exciton lines labelled A, B, C, and D chosen in a 1 meV wide spectral range after the laser was finely tuned to optimize their average intensity. Table I gives the exciton parameters relevant to the study of the exciton spin, namely the fine structure splitting δ_1 and the angle φ_0 of the low-energy eigenaxis (chosen as the $|x\rangle$ state, see Fig. 2(a)), measured by analyzing the linear polarization of the PL under circularly polarized excitation. All excitons exhibit a typical FSS of a few tens of μ eV with eigenaxes which are significantly tilted from the $\langle 110 \rangle$ crystallographic axes. It is therefore crucial to investigate the properties of a single QD to avoid averaging effects making the analysis less convincing [17]. Since these splittings are much larger than the exciton homogeneous linewidth $\hbar/\tau_r < 1$ μ eV, the optical orientation of excitons photocreated with σ^+ polarization averages to zero due to quantum beats [8] as shown in the inset of Fig. 1(c). This contrasts with the case of charged excitons [9, 20].

We first consider the optical alignment of excitons. The excitation linear polarization was set parallel ($\theta = 0, \pi/2$) or at 45° ($\theta = \pm\pi/4$) to the $\langle 110 \rangle$ axes in order to create an exciton either in one of its eigenstates $|x\rangle$, $|y\rangle$ or as a coherent superposition $(|x\rangle \pm |y\rangle)/\sqrt{2}$. As the QD eigenaxes were found tilted from the $\langle 110 \rangle$ directions (see Tab. I) our choice of polarizer directions was not optimal but nevertheless sufficient to reveal strong exciton dipole alignment. The PL linear polarization was analyzed with a polarizer making an angle ϕ with the $[1\bar{1}0]$ direction (see Fig. 2(a)). Two series of spectra obtained by varying ϕ are presented in Fig. 2(b) for two settings of the polarizer, and the degree of linear polarization of exciton B is plotted in Fig. 2(c). All excitonic lines exhibit strong intensity oscillations as a function of the analyzer angle, with maxima and minima corresponding to their eigenaxes, and, remarkably with an amplitude which depends on the excitation polarization. For exciton B which has its eigenaxes tilted by only 7° from the $\langle 110 \rangle$ axes, this effect is particularly pronounced. The oscillation contrast reaches $\sim 90\%$ for $\theta = 0, \pi/2$, namely when the exciton dipole is almost oriented along a QD eigenaxis, but falls down below $\sim 20\%$ when the initial exciton is aligned at 45° ($\theta = \pm\pi/4$). This demonstrates a very efficient optical alignment that can be quantitatively discussed by plotting the degree of linear polarization $P_l(\phi) = (I_\phi - I_{\phi+\pi/2})/(I_\phi + I_{\phi+\pi/2})$ where I_ϕ is the integrated PL intensity of the exciton line for the analyzer at ϕ (see Fig. 2(c)). By treating the

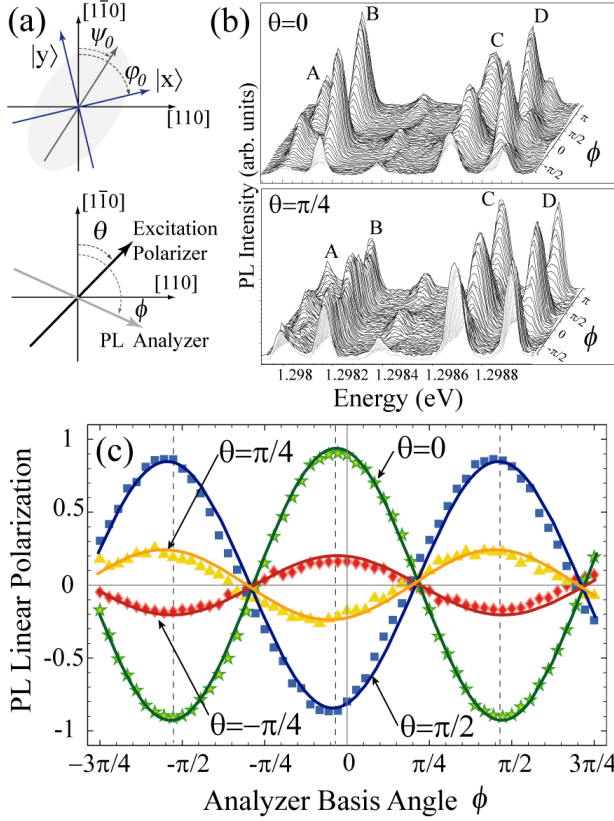


FIG. 2: (a) Scheme of the QD eigenaxis φ_0 and dichroism ψ_0 angles (top) and settings of polarizers (bottom) with respect to the $[1\bar{1}0]$ direction. (b) μ -PL spectra measured for different PL linear analyzer angles ϕ and for two angles θ of the excitation polarization. (c) Linear polarization $P_l(\phi)$ of exciton B (see text), for different linear polarizations θ of the excitation. Solid lines are theoretical fit made with the same set of parameters.

bright exciton as a pseudo-spin 1/2 experiencing an effective in-plane magnetic field $\Omega_{exc.} = \delta_1/\hbar$ [8, 21], we can straightforwardly derive in the density matrix formalism an analytical expression for $P_l(\phi)$:

$$P_l(\phi) = \eta P_l^0 \frac{(2 + \nu^2) \cos 2(\phi - \theta) + \nu^2 \cos 2(\phi + \theta - 2\varphi_0)}{2(1 + \nu^2)} \quad (1)$$

where P_l^0 is the photo-generated initial polarization, $\eta = \tau_s/(\tau_s + \tau_r)$ with τ_s the exciton spin relaxation time [22] and τ_r the exciton lifetime, and $\nu = \eta\Omega_{exc.}\tau_r$. In Eq. 1 we have neglected the effect of exciton thermalization within the bright doublet which would contribute an additional term [23]. Indeed, as a result of the observed large degree of polarization we necessarily have $\eta > 0.92$ so that the thermalization term contributes less than 1% at $T=2$ K. Yet, to explain the small asymmetry of the polarization amplitude between $\theta = 0$ (0.91) and $\theta = \pi/2$ (0.87), as well as a small shift of polarization maxima for $\theta = \pm\pi/4$ it is necessary to include linear dichroism of the

bright exciton due to the reduced symmetry of the QD, which has been modeled by assuming heavy-hole to light-hole mixing [24, 25, 26, 27]. The $|\pm 1\rangle$ bright exciton states which become elliptically polarized are written as $|\pm 1\rangle = \sqrt{1 - \beta^2} |\mp \frac{1}{2}, \pm \frac{3}{2}\rangle + \beta e^{\pm 2i\psi_0} |\mp \frac{1}{2}, \mp \frac{1}{2}\rangle$ with $\beta > 0$ the fraction of light-hole exciton and ψ_0 the angle of dichroism main axis with respect to $[1\bar{1}0]$ (see Fig. 2(a)). As recently discussed for CdTe QDs [26], this angle may differ greatly from the exchange eigenaxis orientation ϕ_0 . The resulting changes of optical selection rules give an additional term in Eq. 1 which to the first order in β can be written:

$$\Delta P_l(\phi) = -\frac{2}{\sqrt{3}}\beta \cos 2(\phi - \psi_0) \quad (2)$$

This theoretical model provides a very good agreement with the experimental results as shown for exciton B in Fig. 2(c) by taking $\beta = 0.04$, $\psi_0 = 93^\circ$, $P_l^0 = 0.95$, $\tau_r = 0.85$ ns, $\tau_s = 20$ ns. We can therefore conclude that (i) the exciton spin can be optically written with a very high fidelity under phonon assisted excitation and (ii) the exciton spin relaxation time is at least one order of magnitude longer than its radiative lifetime.

To investigate further the exciton spin relaxation along directions different from the QD eigenaxes, a magnetic field B_z was applied parallel to the optical axis. The latter is described below by the corresponding Larmor precession angular frequency $\Omega_z = g_{exc.}\mu_B B_z/\hbar$, where $g_{exc.}$ is the exciton g factor and μ_B the Bohr magneton. The total field $\vec{\Omega} = (\Omega_{exc.}, 0, \Omega_z)$, about which the initial exciton spin \vec{S}_0 precesses, becomes oblique with respect to both the optical axis and the QD eigenaxes as sketched in the Poincaré's sphere representation (see Fig. 3(a)). Since the average exciton spin acquires a finite projection on the $|x\rangle$ or $|y\rangle$ states, this gives rise to the phenomenon of polarization conversion previously studied in QWs or QD ensembles [8, 14, 16, 17, 21]. For an individual QD, this effect is spectacular because the broadening due to inhomogeneous distribution of anisotropic splittings δ_1 is suppressed. Note however that polarization conversion would still occur in case of a mixed state (e.g. resulting from fast decoherence of the spin component perpendicular to $\vec{\Omega}$) as long as the relaxation of the spin component parallel to $\vec{\Omega}$ is negligible. The exciton coherence really manifests itself in cw experiments only through the polarization conversion in the direction orthogonal both to $\vec{\Omega}$ and \vec{S}_0 (sine term in Eq. 3 given below), which is however much weaker because of the multiple periods of precession during the exciton lifetime [16].

Experimentally we have observed both linear-to-circular (L2C) and circular-to-linear (C2L) conversion for all the excitonic lines in Fig. 1. Here we focus on the C2L conversion which has been investigated by measuring the linear polarization of individual lines for different angles ϕ of the analyzer under a fixed σ^+ polarized excitation, while the magnetic field was varied from -0.5 T to +0.5 T

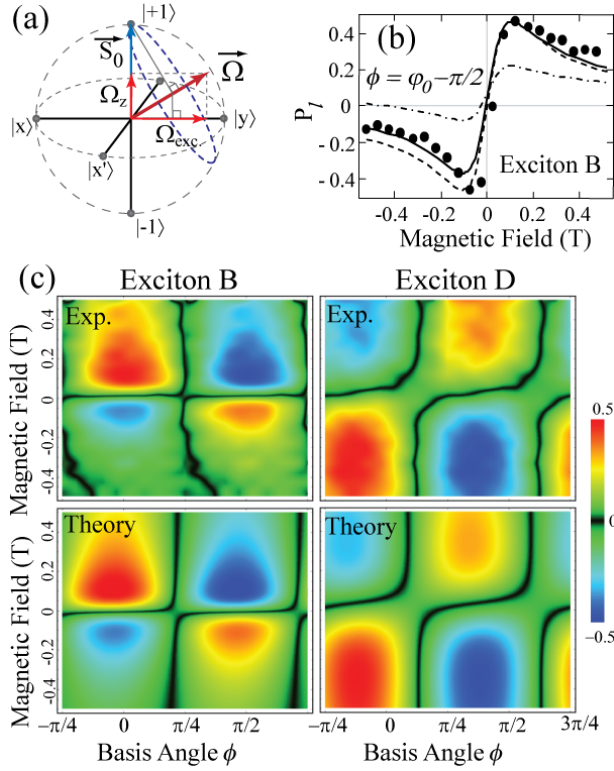


FIG. 3: (a) Scheme of the exciton spin precession on the Poincaré sphere in presence of an external magnetic field Ω_z . (b) C2L conversion of polarization for exciton B. Experimental points (dots), theoretical model (solid line: $\tau_s = 10\tau_r$, $\beta = 0.04$, dashed line: $\tau_s = 10\tau_r$, $\beta = 0$, dot-dashed line: $\tau_s = 0.5\tau_r$, $\beta = 0.04$). (c) Contour-plot of the degree of linear polarization as a function of the magnetic field and angle ϕ of analysis for excitons B and D (top), and theoretical simulation (bottom). See text for details.

with a 50 mT step. The results for exciton B and D are displayed in Fig. 3. C2L conversion manifests itself by the development of a strong linear polarization along the QD eigenaxes ($\phi = \varphi_0 [\pi/2]$), which depends almost anti-symmetrically on B_z and presents clear maxima for finite values of the field (at about ± 100 mT for exciton B, see Fig. 3(b)). Remarkably, this maximum reaches a value 0.47 which is close to the theoretical upper limit (0.5) achieved when $\Omega_z = \Omega_{exc}$, so that the precession vector $\vec{\Omega}$ lies at 45° between $|+1\rangle$ and $|x\rangle$ spin states. For a quantitative analysis, we calculated the field-induced degree of linear polarization for an arbitrary angle ϕ by solving the density matrix evolution of the exciton spin, and obtained the following analytical expression:

$$P_L(\phi) = \sqrt{1 - \gamma^2} \frac{\eta \nu P_c^0}{1 + \nu^2} [\sin 2(\phi - \varphi_0) - \gamma \nu \cos 2(\phi - \varphi_0)] \quad (3)$$

where P_c^0 is the photo-generated initial circular polarization, $\gamma = \Omega_z/\Omega$, and now $\nu = \eta\Omega\tau_r$. As for optical alignment experiments, the spin thermalization could be

neglected. The observed departure from a perfect anti-symmetrical dependence on B_z (predicted by Eq. 3) can again be explained by the linear dichroism of the excitons given by Eq. 2 as shown in Fig. 3(b). For an overall comparison with the model, the linear polarization as a function of magnetic field and analysis angle ϕ has been plotted using a color scale (see Fig. 3(c)). Note for a better rendering (“only” 19 angles and 21 field values were measured) an interpolation has been performed. These 2D plots reveal clearly how the basis of maximum C2L conversion rotates from $\phi = \varphi_0 + \pi/4 [\pi/2]$ to $\phi = \varphi_0 [\pi/2]$, when the magnetic field strength increases, in agreement with both righthand terms in Eq. 3. By taking into account the exciton parameters of Tab. I, we could reproduce reasonably well the experimental contour-plots with Eqs. 2,3 as illustrated in the bottom part of Fig. 3(c). For both excitons B and D, we used $g_{exc.} = 2.5$, $\tau_r = 0.85$ ns, $\tau_s = 10$ ns, but different values of valence band mixing, namely $\beta = 0.04$ (0.1) and $\psi_0 = 93^\circ$ (90°) for exciton B (D) respectively.

In summary, optical alignment of exciton spin along each of its linearly polarized eigenstates was demonstrated in zero field, whereas a very efficient conversion of polarization from a circularly- to linearly-polarized exciton was achieved by applying a longitudinal magnetic field. Our results, discussed within the framework of a theoretical model including valence band mixing effects on exciton optical selection rules, prove straightforwardly that the exciton spin of an isolated QD has a very long relaxation time in comparison to the exciton radiative lifetime, a conclusion which is of fundamental importance for the development of QD-based sources emitting polarization entangled photon pairs.

One of us (K.K.) was supported by the European network of excellence SANDIE and by the Foundation for Polish Science (START programme). This work has been partially supported by the Polish Ministry of Science and Higher Education (Grants financed in 2007-2010).

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